25. Monoclinic double selenates of the manganese group.
'Proc. Roy. Soc.,' A, vol. 101, p. 225 (1922).
26. Monoclinic double selenates of the cadmium group.
'Proc. Roy. Soc.,' A, vol. 101, p. 245 (1922).

MEMOIRS CONCERNING NEW INSTRUMENTS FOR THE RESEARCH.

1. An instrument for grinding section-plates and prisms of crystals of artificial preparations accurately in the desired directions.
'Phil. Trans.,' A, vol. 185, p. 887 (1895); 'Z. f. Kryst.,' vol. 24, p. 433 (1895).
2. An instrument of precision for producing monochromatic light of any desired wavelength, and its use in the investigation of the optical properties of crystals.
'Phil. Trans.,' A, vol. 185, p. 913 (1895); 'Z. f. Kryst.,' vol. 24, p. 455 (1895).
3. An instrument for cutting, grinding and polishing section plates and prisms of mineral or other crystals accurately in the desired directions.
'Proc. Roy. Soc.,' A, vol. 57, p. 324 (1895); 'Z. f. Kryst.,' vol. 25, p. 79 (1896).
4. A compensated interference dilatometer.
'Phil. Trans.,' A, vol. 191, p. 313 (1898); 'Z. f. Kryst.,' vol. 30, p. 529 (1899).
5. Verbesserungen an den Apparate zum Schneiden, Schleifen, und Poliren genau orientirter Krystallplatten. 'Zeitschr. für Kryst.,' vol. 31, p. 458 (1899).
6. The elasmometer, a new interferential form of elasticity apparatus.
'Phil. Trans.,' A, vol. 202, p. 143 (1904); 'Z. f. Kryst.,' vol. 39, p. 321 (1904).

Magnetism and Atomic Structure.—II. The Constitution of the Hydrogen-Palladium System and other similar Systems.

By A. E. OXLEY, M.A., D.Sc., F.Inst.P.

(Communicated by A. W. Porter, F.R.S. Received August 17, 1921.)

1. INTRODUCTION.

The present work is a continuation of that published in 'Phil. Trans., A, vol. 214, pp. 109–146 (1914); A, vol. 215, pp. 79–103 (1915); A, vol. 220, pp. 247–289 (1920); 'Roy. Soc. Proc.,' A, vol. 95, p. 58 (1918), and A, vol. 98, p. 264 (1921). It was believed that a magnetic examination of the properties of palladium black which had been charged with hydrogen would enable us to decide which of the proposed constitutions is the correct one. If the hydrogen were in the atomic state and *free,* one might expect that the system would be more magnetic than uncharged palladium black, whether the assumed constitution of the hydrogen atom be that of Bohr* or that proposed by the author.* If the contained hydrogen were in the molecular form, either in the state of a gas or condensed to a liquid, since gaseous and liquid hydrogen are both diamagnetic,† we should expect a diminution of the magnetic property of the system by a small amount, depending on the minute diamagnetic susceptibility of molecular hydrogen and the amount of gas occluded. Lastly, if the hydrogen enters into combination with the palladium, or is associated with the palladium so as to form a type of loose chemical compound, we should, in the light of previous knowledge of the effects of chemical combination on magnetic susceptibility, expect a change depending on the nature of the compound formed.

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* Sommerfeld, 'Atombau u. Spectrallinien,' p. 239, *et seq.*

It is impossible here to go into details regarding the vast amount of work which has been undertaken to solve the problem of gaseous occlusion. Reference is made to the General Discussion held by the Faraday Society on the subject.‡ It will be convenient to summarise here the diverse views at present held as to the nature of the complex process of occlusion.§ These have been given by A. W. Porter|| under the following six heads :—

(1) Chemical combination of the gas with the metal.

(2) Simple solid solution, either separately or in conjunction with (1).

(3) Two solid solutions in contiguous phases.

(4) Solution accompanied by surface adsorption.

(5) Surface condensation under molecular forces unaccompanied by solution.

(6) Simple inclusion of the gas in the interstices of the metal.

2. EXPERIMENTAL DETERMINATION OF THE MAGNETIC PROPERTIES OF PALLADIUM TREATED WITH HYDROGEN.

The apparatus used was a modification of that described in 'Phil. Trans.,' A, vol. 214, p. 112 (1914), with the following differences. On account of the high susceptibility of pure palladium compared with that of the substances which were investigated in the earlier work, it was decided to abandon the delicate bifilar suspension mirror. Instead, a tele-microscope, focussed on

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* 'Nature,' vol. 105, p. 327 (1920). It is not inconceivable that the electron in the *free* hydrogen atom may move up into an orbit in the nuclear plane, a modification determined by conditions of symmetry.

† T. Soné, 'Science Reports, Tohoku,' vol. 8, p. 115, 1919 (for gaseous H $\chi = -19{\cdot}8 \times 10^{-7}$) ; K. Onnes, 'Proc. Amsterdam Acad.,' vol. 14, p. 121, 1911 (for liquid H $\chi = -27 \times 10^{-7}$).

‡ 'Trans. Faraday Soc.,' vol. 14, Part III (1919).

§ In what follows, "occlusion" is used in a general way to include the rapid process of adsorption and the slower process of absorption or diffusion. McBain uses the term *sorption* in the same sense.

|| *Ibid.*, p. 192.

the support A′* carrying the specimen, was used. A nul method was adopted, the force exerted on the specimen being counterbalanced by the torsion of the phosphor-bronze strip and read off on the torsion head.

The sighting arrangement is shown in fig. 1. To prevent disturbance on the suspended system due to air currents, a brass cylinder, cut so as to fit over the pole-pieces and slotted so that it could be readily placed in position when the specimen was attached to the point A of the torsion arm, was provided. The setting of the torsion head was adjusted so that the left side of the vertical support carrying the specimen was coincident with the setting line B, as viewed through the tele-microscope.

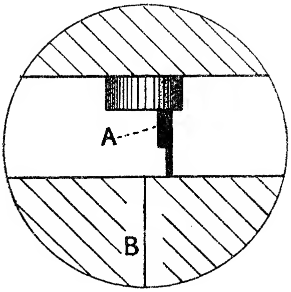

Fig. 1.

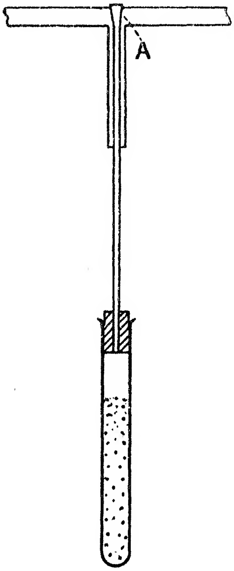

Fig. 2.

The phial in which the specimen was placed, shown in fig. 2, consisted of a thin-walled glass tube 0·63 cm. diameter and 4·75 cm. long.

Preparation of the Hydrogen-Palladium Specimens.

The author is indebted to Dr. F. P. Burt for the loan of 2 grms. of palladium black. This was divided into two approximately equal parts, one of these being charged with hydrogen in the apparatus shown in fig. 3. The activity of the specimens was considerable, and the particles of charged palladium showed vivid flashing when they struck the walls of the tube.

The other half of the palladium was not treated. Its susceptibility was practically identical with that obtained by previous investigators for pure palladium, indicating that this portion was free from occluded hydrogen or combined oxygen, at least to any measurable extent. Subsequent experiments, in which this sample was treated with hydrogen, heated to over 200° C. and the hydrogen pumped off, gave products whose susceptibility did not differ from that of the untreated one by more than 1 per cent.

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* See fig. 2 and 'Phil. Trans.,' *loc. cit.*, fig. 2.

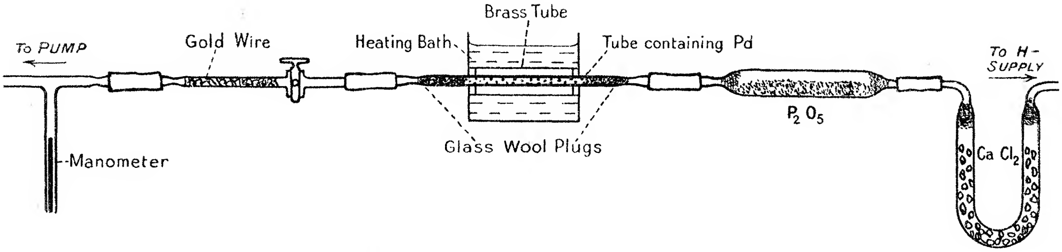

FIG. 3.

Measurement of the Specific Susceptibility (χ) *of the Specimens.*

The equation for calculating this quantity is*

$$\chi = \frac{1}{M} \cdot \left[\chi_a \cdot M_a + (\chi_\omega \cdot m_\omega - \chi_a \cdot m_a) \cdot \frac{\vartheta - \vartheta_1}{\vartheta_2 - \vartheta_1}\right] \ldots, \qquad (1)$$

where ϑ_1 is the torsion due to phial and contained air,
ϑ_2 is the torsion due to phial and contained water,
ϑ is the torsion due to phial and contained specimen,
χ_a is the specific susceptibility of air,
χ_ω is the specific susceptibility of water,
χ is the specific susceptibility of specimen,
M is the mass of specimen,
M_a is the mass of air filling the same volume as the specimen,
m_ω is the mass of water,
m_a is the mass of air filling the same volume as the water.

The values of χ_ω and χ_a were taken as $-7{\cdot}25 \times 10^{-7}$ and 210×10^{-7} (at 20° C.) respectively.

In the following Tables, Z_0 is the zero of the torsion head reading, Z_1 the reading when the phial had been brought back to its initial position, ϑ, ϑ_1 and ϑ_2 the torsion necessary to balance the force on the phial when the latter was filled with the substance, air and water respectively. All the experiments (see Table I) were made at room temperature 17°–19° C., and each of the readings recorded is the mean of several determinations which agreed to within 2 per cent.

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* *Cf.* 'Phil. Trans.,' A, vol 214, p. 114 (1914).

Table I.

Experiment.	Weight.	Z_0.	Z_1.	Z_0.	ϑ.	ϑ_1.	ϑ_2.	H (gauss).	Remarks.
I—(*a*) Pd—black charged with hydrogen	grm. 0·838	260	3 × 360 − 338	260	1158	—	—	3200	
	0·838	258	3 × 360 − 323	258	1145	—	—	3200	Phial replaced.
(*b*) Pd—black, uncharged	0·933	253	4 × 360 − 217	252	1766	—	—	3200	Torsion head reset.
	0·933	230	4 × 360 − 199	230	1769	—	—	3200	Phial replaced.
	0·933	230	4 × 360 − 212	230	1782	—	—	3200	,, ,,
(*c*) Repeated (*a*) after 24 hours,	0·838	232	3 × 360 − 122	232	1330	—	—	3200	After 24 hours' exposure to air.
+ 14 hours	0·838	230	3 × 360 − 204	231	1414	—	—	3200	,, 38 ,, ,, ,,
(*d*) Phial filled with air	2·052	230	1 × 360 − 225	230	—	356	—	3200	
	2·052	230	1 × 360 − 220	230	—	350	—	3200	Phial replaced.
(*e*) Phial filled with water	3·199	228	23	228	—	—	155	3200	
	3·199	229	13	229	—	—	144	3200	,, ,,
II—(*f*) Pd—black charged with	0·809	29	328	29	299	—	—	2000	Torsion head reset.
hydrogen	0·809	29	326	29	297	—	—	2000	Phial replaced.
(*g*) Pd—black, uncharged	0·829	28	382	29	354	—	—	2000	
	0·829	28	374	28	346	—	—	2000	,, ,,
	0·829	28	371	28	343	—	—	2000	,, ,,
(*h*) Phial filled with air	2·056	28	114	28	—	86	—	2000	
(*i*) Phial filled with water	3·135	29	76	29	—	—	47	2000	

Table II.

Experiment.	Weight.	Z_0.	Z_1.	Z_0.	ϑ.	ϑ_1.	ϑ_2.	H (gauss).	Remarks.
V.—(*j*) Pd—black charged with	grm. 0·658	26	162	26	136	—	—	2000	
hydrogen, sealed tube	0·658	27	164	27	137	—	—	2000	Phial replaced.
	0·658	28	164	28	136	—	—	2000	,, ,,
(*k*) Sample (*j*) after the	0·658	28	278	28	250	—	—	2000	
hydrogen had been	0·658	28	275	28	247	—	—	2000	,, ,,
pumped off	0·658	28	272	28	244	—	—	2000	,, ,,
(*l*) Phial filled with air	6·130	29	134	29	—	105	—	2000	
	6·130	29	134	29	—	105	—	2000	,, ,,
(*m*) Phial filled with water......	7·270	29	105	29	—	—	76	2000	
	7·270	29	105	29	—	—	76	2000	,, ,,

A third and fourth series of experiments similar to I and II have been made. The results agreed with those given above, and showed that the deflections obtained with the "activated" palladium were always less than those for the pure palladium.

The Hydrogen-Content of the Charged Samples.

At the conclusion of the second, third, and fourth series of tests, the palladium was transferred to a glass tube, sealed at one end, and connected through the tube containing gold wire, to a manometer and Topler pump. The palladium was heated, and the hydrogen given off collected and measured. All the palladium samples had been subjected to hydrogen at a pressure a little greater than atmospheric for an hour and three quarters, and allowed to cool in contact with the gas. From 0·83 grm. of palladium black, the volume of gas collected was 20, 17, and 15 c.c. in the three cases. The occlusion capacity is usually referred to palladium in the crystalline state, whose density is 11·4, so that, at the close of the magnetic experiments, 0·08 c.c. of palladium contained 20, 17, and 15 c.c. of hydrogen, or the palladium contained 250, 212, and 188 times its own volume of hydrogen. This is a small amount, but the loss is due in the main to the combination of the active hydrogen with the oxygen of the air, as evidenced by the dampness of the specimens and the change in appearance from a metallic lustre to coal-black. A further loss resulted from mechanical disturbance during the transference to and from the phial and charging and discharging tubes. The flashing which took place and the heat evolved were noticeable. An estimate of the hydrogen loss through the formation of water was made with the last of the above specimens. This was weighed in the damp state and in the dry state after the hydrogen had been pumped off. A difference of 0·097 grm. was found.* This corresponds to a loss of about 120 c.c. of hydrogen, so that, in the initial stage, the samples would probably contain 300–350 times their own volume of hydrogen, the loss due to mechanical disturbance being an uncertain amount.

Experiments with Sealed Tube.

An attempt was made to prevent loss of hydrogen due to chemical combination with the oxygen of the air as well as that due to mechanical disturbance of the specimen.

The phial used, shown in fig. 4, consisted of an outer glass tube, 9 mm.

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* Magnetically, this amount of water would have no effect on the measurements, the specific susceptibility of water being $-7{\cdot}25 \times 10^{-7}$. Further, the mass effect does not come in because the specimens were weighed before exposure to the air took place.

diameter and 7·8 cm. long. The palladium specimen was contained in a smaller tube, 6 mm. diameter and 7·3 cm. long. This tube was used during the charging operation (see fig. 3). The pure palladium weighed 0·658 grm. This was placed in the tube, two glass-wool plugs being inserted as before, and the tube drawn out so as to form constrictions beyond the plugs. The charging was continued for 3 hours, and the palladium allowed to cool in the hydrogen for 2 hours. The tube was cut at the two constrictions, and a small dab of white wax immediately placed over the drawn-out open ends. This sealed tube was placed in the larger tube and suspended from the torsion arm at A. A series of readings was now made with the specimen in the double-walled phial (see Table II). Afterwards the phial was opened, and the palladium tube placed inside a wide tube, which was connected through a tube of gold wire with the manometer and Topler pump. The apparatus was then exhausted. By applying a small flame to the outer tube, the speck of wax was softened and the palladium tube opened to the vacuum. Afterwards the temperature of the palladium tube was raised to about 200° C. and the whole of the hydrogen pumped off. The volume of gas collected was 49 c.c. About 1 c.c. of this was unoccluded hydrogen, and therefore 0·66 grm. of palladium black contained 48 c.c. of hydrogen. Crystalline palladium has a density of 11·4, and, if the 0·66 grm. of palladium black were compressed to this density, its volume would be 0·06 c.c. nearly. This volume occludes 48 c.c. of hydrogen, and therefore one volume of crystalline palladium subjected to the same treatment would contain 48/0·06, or 800 c.c. of hydrogen.

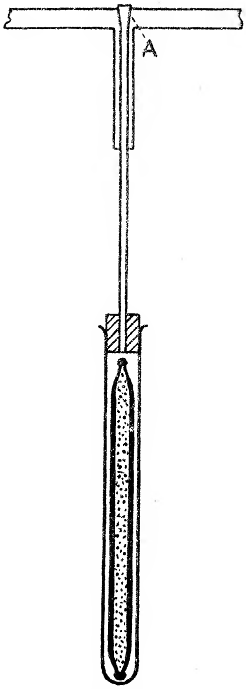

Fig. 4.

When all the hydrogen had been pumped off, the palladium was allowed to cool in the vacuum. Air was next gently admitted, so as not to disturb the palladium, and a speck of wax again placed on the unsealed end of the tube. (It was found that the difference in weight between the tube, as originally sealed and as now sealed, was 0·026 grm., this being the excess of white wax in the former case over that in the latter. A subsidiary experiment showed that the wax was slightly paramagnetic, but the mass here involved would not affect the resulting deflection by more than one or two scale divisions, and, in any case, the difference due to this cause was purposely made so as to give the slight increased deflection to the activated sample whose magnetic property, as indicated by the earlier determination, was less than that of pure palladium.)

The palladium tube was replaced in the phial and a series of readings taken as before (see Table II). During the above experiment it was observed that the active palladium preserved its metallic lustre, and when the tube containing it was rotated there was no adhesion of the particles to its walls, as was noticed in the previous specimens.

Calculation of the Susceptibilities of the Specimens.

From equation (1) the specific susceptibilities of the active and inactive palladium specimens are:—

$$\chi_{\text{inactive}} = +64{\cdot}6\,(7)\times 10^{-7},$$

$$\chi_{\text{active}} = 36{\cdot}5\times\underbrace{10^{-7},\ 44{\cdot}6\times}_{24\text{ hours},}\ \underbrace{10^{-7},\ 48{\cdot}5\times}_{+14\text{ hours.}}10^{-7}\text{ after}$$

It appeared that the specific susceptibility was gradually returning to the value for the inactive specimen as the occluded hydrogen escaped through standing in contact with air. Fig. 5 shows the rate of recovery with time.*

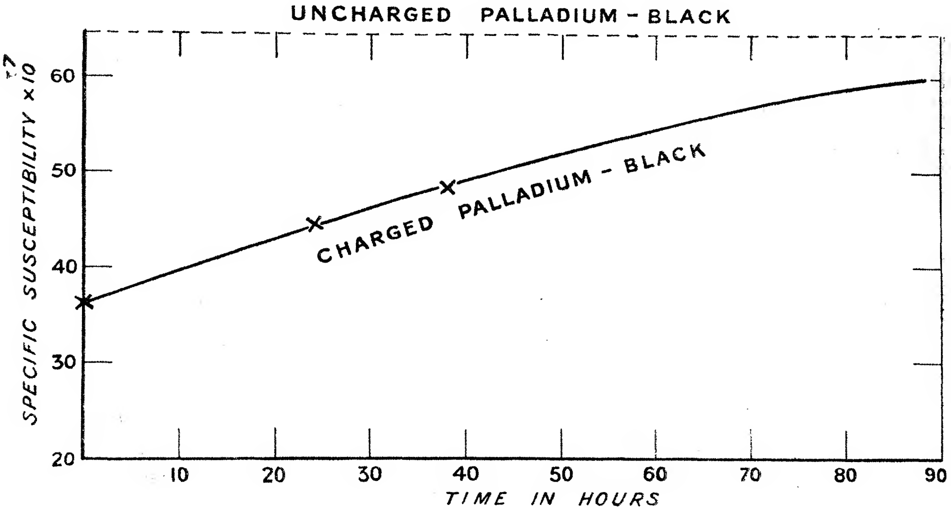

Fig. 5.

From the second series of experiments, the specific susceptibilities are:—

$$\chi_{\text{inactive}} = +64{\cdot}0\times 10^{-7},$$

$$\chi_{\text{active}} = +53{\cdot}4\times 10^{-7}, \qquad +51{\cdot}0\times 10^{-7}.$$

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* See footnote, p. 269.

The active specimen was again less paramagnetic than the inactive one by approximately 20 per cent., a value less than that obtained in the first series. The third and fourth series gave results of the same order as the second series.

The fifth series, as was to be expected, gave the greatest reduction, viz. :—

$$\chi_{\text{inactive}} = +64{\cdot}3 \times 10^{-7},^{*}$$

$$\chi_{\text{active}} = +14{\cdot}7 \times 10^{-7},$$

a reduction of 75 per cent.

3. INTERPRETATION OF THE EXPERIMENTAL RESULTS.

The hydrogen atom contains a single unbalanced electron, and, assuming that the nucleus, even if it rotates, contributes nothing to the magnetic properties, on account of its minute size, we should expect that *free* hydrogen atoms occluded in the palladium would involve an increase in the specific susceptibility. The experimental results described above therefore indicate that the hydrogen occluded by palladium is not in the *free* atomic state.

Molecular hydrogen, both in the gaseous and liquid state, is diamagnetic, and the existence of the occluded hydrogen in either of these states would therefore imply a smaller specific susceptibility for the active palladium. Such molecular hydrogen would not combine with the palladium. Therefore, knowing the specific susceptibility of hydrogen and the mass occluded, we can calculate the amount by which the specific susceptibility of the palladium would be reduced. Now, palladium of normal density 11·4 occludes 640 times its volume of hydrogen. Taking the mass of palladium black used in the earlier experiments as approximately 0·9 grm., and its volume as 1 c.c., the volume of hydrogen occluded by it should be of the order 70 c.c. The mass of this is 0·0063 grm. Taking the specific susceptibility of hydrogen as -30×10^{-7}, this occluded hydrogen would reduce the specific susceptibility of the palladium by $0{\cdot}19 \times 10^{-7}$, which is quite insufficient to account for the observed difference. Hence the occluded hydrogen is not *gaseous molecular* hydrogen or *condensed molecular* hydrogen in a state of mere solid solution or dissolved in the metal.

The magnetic property of a metallic compound is usually different from that of the components taken together. Thus iron and nickel are ferromagnetic, carbon monoxide is diamagnetic. The two carbonyls of iron and nickel, $Fe(CO)_5$ and $Ni(CO)_4$, are diamagnetic,† having susceptibilities comparable with that of water. It is possible that something of this nature takes place when palladium occludes hydrogen. If a compound of palladium and hydrogen were formed which is diamagnetic or, at any rate, much less magnetic than pure palladium, the specific susceptibility of the hydrogen-palladium system might be reduced 50 per cent. or more below that of pure palladium.

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* Honda, 'Ann. d. Phys.,' vol. 32, p. 1044 (1910), gives $+61{\cdot}2 \times 10^{-7}$, other values ranging down to $+58 \times 10^{-7}$ have been found.

† A. E. Oxley, 'Proc. Cam. Phil. Soc.,' vol. 16, p. 102 (1911).

One grm. of palladium contains $5{\cdot}7 \times 10^{21}$ atoms; 1 grm. of the palladium black, whose density is approximately 1, will occlude about 64 c.c. of hydrogen, which contains $3{\cdot}4 \times 10^{21}$ atoms of hydrogen. The ratio of the number of hydrogen atoms to the number of palladium atoms in 1 grm. of the palladium black is $3{\cdot}4/5{\cdot}7$ or 60 per cent. It is curious that the proportion of hydrogen atoms is a little greater than that of half the palladium atoms. We may perhaps suppose that a *loose* type of chemical compound is formed, loose because of the known great instability of the system and the readiness with which the occluded hydrogen is given off. Each hydrogen atom may be pictured as oscillating between two palladium atoms with which it is alternately in combination. The composition of the system would then correspond to that of the supposed compound Pd_2H, a view advocated by Troost, Hautefeuille, and by Wolf. It is possible for palladium to occlude still greater quantities of hydrogen, when the external pressure is increased, and in such cases there may be a tendency for a loose compound of the type PdH to form, or the excess hydrogen may form a solid solution in the system beyond this concentration.*

The magnetic results given in section (2) indicate a considerable drop in the susceptibility, varying from 70 per cent. in the fifth series, 40 per cent. in the first series, to 25 or 20 per cent. in the other series, and it is considered that the formation of an unstable compound or perhaps even a mere wandering of the hydrogen atom over the "surfaces" of the palladium atoms, to which the former temporarily attach themselves, is what actually occurs.

Now suppose that when a hydrogen atom is temporarily attached to a palladium atom the compound PdH formed is diamagnetic. At any moment only one-half the palladium atoms would show their normal magnetic property. Assuming a structure for the hydrogen atom like that suggested by the author† it is possible to picture the combination as shown in fig. 6, where the electron of the hydrogen atom has gone into the outer shell of electrons of the palladium atom. The PdH system would thus consist of two nuclei, those of normal palladium and hydrogen, with a system of electrons very similar to that of silver, whose atomic number is one greater than palladium. Reasons have been given above for supposing that the magnetic property of elements depends entirely on their electronic configuration, and therefore we should expect the temporary compound PdH to be, like silver, diamagnetic.

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* Ramsay, Mond and Shields, 'Phil. Trans.,' A, vol. 191, p. 105 (1898), indicate the possibility of the formation of a hydride Pd_3H_2 for large hydrogen contents.

† 'Roy. Soc. Proc.,' A, vol. 98, p. 264 (1921), and 'Nature,' vol. 105, p. 327 (1920).

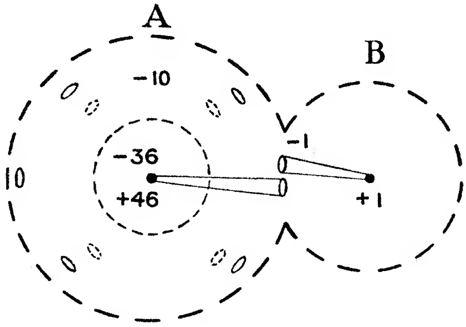

Fig. 6.

The specific susceptibity of silver is $-1{\cdot}86 \times 10^{-7}$, and, if half the electronic systems are similar to those of silver, their contribution to the magnetic property of 1 grm. of active palladium would be $-0{\cdot}93 \times 10^{-7}$, a quantity which is negligible compared with the susceptibility of palladium. Therefore the susceptibility of the palladium would be reduced by 50 per cent. As the occluded hydrogen escaped owing to mechanical disturbance, or combined with the oxygen of the air, the susceptibility would gradually rise until it reached the value for pure palladium (*cf.* fig. 5).

The electrical conductivity of palladium may be attributed to a tendency of the electrons in the outer shell of the atom to wander. It is conceivable that the attachment of a hydrogen atom in the manner indicated in fig. 6 would tend to counteract this, the hydrogen atom acting with regard to the nomad as a sort of anchor. Hence the electrical conductivity of the hydrogen-palladium system would fall off as the number of loose PdH units increased.* This agrees with Wolf's experiments.

The process of occlusion of hydrogen in crystalline palladium is a difficult one to understand. Holt and Andrew† have shown that if sufficient time is allowed, the capacity for occlusion is the same for crystalline and amorphous palladium. The atomic volume of palladium is not very different from that of hydrogen, and therefore the expansion of a palladium wire by about 15 per cent., as observed by Graham,[*] the wire containing 600 times its own volume of hydrogen, is very small. It seems necessary to imagine that the crystalline lattice of palladium is hollow, like the diamond lattice.[†] If the hollowness is the determining factor in occlusion, this may throw some light on the atomic nature of the occluded gas, since molecules of hydrogen would not find a cavity large enough to accommodate them. It is important to note that the lattices of both palladium and silver are face-centred cubic. In the more open structures of the amorphous palladium, cavities must exist large enough to receive numbers of hydrogen molecules. Supposing these have entered, let us assume that there is an appreciable attraction between the Pd and H atoms which is comparable with that between two H atoms. Under this differential attraction and favourable collisions, a hydrogen molecule may be broken up and one hydrogen atom find itself in a hollow in the palladium lattice. This process may be repeated with the gradual diffusion of the hydrogen atoms through the palladium accompanied by the break up of the lattice. We know that diffusion of this kind does take place and we must assume that the thermal oscillations of the Pd atoms are sufficient to allow a way through for the hydrogen atoms eventually, though the process is very slow. The increased power of occlusion as the temperature of the palladium is raised to 100° C. is evidence of this. The expulsion of the hydrogen produced at 180° C. and above is due to oscillations of larger amplitude which permit the hydrogen atoms to unite and form molecules.

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* Silver is a good conductor, and if its electron system is similar to that of Pd—H we must attribute the difference in the conductivities to the presence (externally) of the H nucleus in the Pd—H system.

† 'Roy. Soc. Proc.,' A, vol. 89, p. 170 (1913); A, vol. 90, p. 226 (1914).

The experiments of Graham,[‡] which indicate that *crystalline* palladium becomes more magnetic when charged with hydrogen, can be explained on the views expressed here.

His deduction of the existence of a magnetic "hydrogenium" is not in any way supported by more recent researches on the magnetic property of hydrogen in combination. Liquid[§] and gaseous[||] hydrogen are diamagnetic, and atomic hydrogen[¶] in all the hydrocarbons is diamagnetic. The free hydrogen atom is paramagnetic, but when it combines with a carbon atom the system is a compensated one. It is possible that each of the four hydrogen atoms in CH_4 holds a pair of electrons in common with the carbon atom. In the diamond, each of the four outer electrons of any atom is one of a pair of electrons held in common between two atoms, and recent X-ray evidence supports the view that weak diffraction centres exist half-way between the carbon nuclei in the diamond. In the amorphous variety this bonding of the carbon atoms is inappreciable; the rigidity is small, but the power of occlusion large. May it not be that this capacity for occlusion is determined by a combination, similar to that which has been prescribed for the Pd—H system, the molecules of the occluded element first becoming dissociated, and finally sharing an electron with the carbon atom? In the case of the diamond, where the electrons of the carbon atoms are already shared among themselves, occlusion would be impossible, even though there are hollows in the diamond lattice.

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[*] 'Journ. Chem. Soc.,' vol. 22, p. 430 (1869).

[†] Sir William Bragg, 'Nature,' vol. 105, p. 79 (1921). (Abstract of Kelvin Lecture.)

[‡] 'Journ. Chem. Soc.,' vol. 22, p. 430 (1869).

[§] Onnes, 'Proc. Amsterdam Acad.,' vol. 14, p. 121 (1911).

[||] Soné, 'Science Reports, Tohoku,' vol. 8, p. 115 (1919).

[¶] Pascal, 'Ann. de Chim. et de Phys.,' vol. 8, p. 19 (1910).

The two varieties of palladium are very similar in their behaviour to those of carbon. Amorphous carbon has a specific susceptibility $-20 \cdot 2 \times 10^{-7}$, diamond $-5 \cdot 0 \times 10^{-7}$, or four times smaller. The specific susceptibility of palladium black is 58×10^{-7}, crystalline palladium 11×10^{-7}, or five times smaller. Crystalline palladium, like diamond, is rigid, because its atoms are held together by electron sharing. The amorphous varieties have no cohesion, but high occlusion capacity. The greater the amount of amorphous palladium, the more rapidly will the hydrogen be occluded, and the expansion which results disintegrates the crystalline grains producing a larger amount of amorphous palladium. It is conceivable that circumstances may arise whereby, through internal strain, minute masses of the amorphous variety may be formed, which are screened off from attack by the hydrogen atoms by crystalline fragments through which the hydrogen diffuses very slowly. The heterogeneous system so formed would have a larger susceptibility than the original crystalline mass in proportion to the amount of such amorphous phase formed. When the hydrogen is driven off by heat, the palladium retracts, as Graham showed, and does so beyond its original dimensions, probably owing to the opportunity given to readjust internal strains which originally existed in the crystalline metal. Such retraction is often accompanied by a reduction of the occlusion capacity when the sample is again treated with hydrogen, and this is what might be expected. The properties of palladium in which the crystalline and amorphous phases co-exist are clearly complex, and a satisfactory solution can only be given when the relative proportions of these phases is determinate.

It has been suggested that, as regards the properties of the outer regions of the atom only, we may look upon any complex atom, *e.g.*, a palladium atom, as consisting of an assemblage of hydrogen atoms.* If a free hydrogen atom attaches itself to such a system as shown in fig. 6, a considerable amount of heat would be evolved. For two hydrogen atoms combining, the thermal evolution is 77,000 calories per gramme.* Mond, Ramsay, and Shields have shown that the heat set free when 1 grm. of hydrogen is occluded in palladium is 6,472 calories. The thermal value of the combination of palladium and hydrogen will be the sum of these numbers, or 83,500 calories per gramme of combined hydrogen. This implies that the Pd—H system, in the absence of excess hydrogen, is exceedingly stable. The presence of other hydrogen atoms, or oxygen (if the system is in contact with air), will make the Pd—H system less stable, and measurable in fact by the thermal change 6,472 calories per gramme of hydrogen, as determined by Mond, Ramsay, and Shields. The view which has often been put forward that the hydrogen is merely condensed as a liquid within the metal, not only gives the low thermal change of 123 calories for the occlusion of 1 grm. of hydrogen, but demands the presence of very stable hydrogen molecules which are inconsistent with the great activity of the hydrogen-palladium system.

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* A. E. Oxley, 'Nature,' vol. 105, p. 327 (1920). Any negative charges necessary to cement the positive charges of the nucleus are not involved here The disintegration of a hydrogen nucleus would depend on the attraction between it and the nuclear electrons; once this had taken place, *e.g.*, by bombardment with α-particles (*cf.* Rutherford, 'Phil. Mag.,' vol. 41, p. 513 (1920)), the H-nucleus would take up an electron and become a neutral H-atom.

4. THEORETICAL APPLICATION OF THE ABOVE RESULTS TO OTHER SYSTEMS.

The formation of an electronic system corresponding to an element of higher atomic number than that of the occluding element, such as those of the palladium-hydrogen and silver systems described above, is suggestive in connection with the change of magnetic properties of other elements containing occluded hydrogen. It has been shown that pure manganese is paramagnetic, its specific susceptibility at 18° C. being $+11 \times 10^{-6}$. When manganese is fused in an atmosphere of hydrogen, or is deposited electrolytically, it is ferro-magnetic, the highest values of χ being of the order $+2000 \times 10^{-6}$. Such manganese contains a certain amount of occluded hydrogen, the abstraction of which causes the manganese to assume its normal paramagnetic susceptibility.†

Now, the atomic number of manganese is 25, that of iron 26. If we suppose that, when hydrogen is occluded in manganese, the electron of the hydrogen atom enters into the outer shell of the manganese atom, an electronic system similar to that of iron would result. As before, the Mn—H system differs from the Fe system, since the former has two nuclei, one of manganese the other of hydrogen, while the latter has the single iron nucleus. We might expect, therefore, that the presence of hydrogen in manganese would tend a make a certain number of the manganese atoms ferro-magnetic, in proportion to the number of hydrogen atoms occluded in the manganese.

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* Langmuir, 'Phil. Mag.,' vol. 27, p. 188 (1914).

† Hadfield, Chéneveau and Geneau, 'Roy. Soc. Proc.,' A, vol. 94, p. 69 (1917); also Weiss and Onnes, 'Comptes Rendus,' vol. 150, p. 686 (1910).

We can calculate the amount of hydrogen which would be necessary to produce the rise of susceptibility referred to above. The saturation moment of 1 grm.-atom of iron is 11×1123,* and the magnetic moment of each atom $11 \times 1123/6{\cdot}06 \times 10^{23}$. Suppose that the hydrogen atoms in 1 grm. of manganese are effective in producing N electronic systems similar to that of the iron atom. The magnetic moment of these will be

$$N \times 11 \times 1123/6{\cdot}06 \times 10^{23}.$$

The rise of susceptibility of the manganese is from $11{\cdot}0 \times 10^{-6}$ to 2200×10^{-6}, or 2189×10^{-6}, for a field strength of 320† gauss. This implies an increase of magnetic moment of $2189 \times 10^{-6} \times 320$ per gramme. Hence

$$N = 2189 \times 10^{-6} \times 320 \times 6{\cdot}06 \times 10^{23}/1123 \times 11 = 3{\cdot}2 \times 10^{19}.$$

This is the number of Mn—H systems present, and is also equal to the number of H atoms per *gramme* of manganese necessary to produce the ferro-magnetism in the manganese. The mass of this hydrogen is $3{\cdot}2 \times 10^{19}/6{\cdot}06 \times 10^{23}$, or $5{\cdot}4 \times 10^{-5}$ grm., and its volume nearly 0·6 c.c. per gramme of manganese. The density of manganese is 7·5, and therefore, according to this calculation, this element would occlude 4·5 times its own volume of hydrogen. This is a reasonable estimate compared with the capacities for occlusion of other elements‡ :—

	Vols.		Vols.
Palladium black	502	Nickel	17·6
Platinum sponge	49·3	Copper	4·5
Gold	46·3	Aluminium	2·7
Iron	19·2	Lead	0·15

In soft iron the coercive force is about 2 and sometimes less; that of cobalt may reach a value as high as 12.§ The fact that iron which has been fused in an atmosphere of hydrogen has a larger coercive force than 2 is suggestive. In this case it is conceivable that the occluded hydrogen atoms thrust their electrons into the outer shells of the iron atoms, and produce a combined electronic system analogous to that of cobalt.

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* Weiss, 'Archives des Sciences Phys. et Nat.,' vol. 31 (1911).

† Hadfield, Chéneveau and Geneau, *loc. cit.*, p. 70, footnote.

[*Note added, January* 8, 1922.—The amount of hydrogen in this specimen was not measured, but Sir Robert Hadfield informs me that in a sample of electrolytic manganese, 45 c.c. of hydrogen were driven off per cubic centimetre of manganese. Kuh ('Thèse,' Zurich, 1911) has obtained a magnetic moment for ferromagnetic manganese twice as large as that recorded above, and this would correspond to 9 c.c. of occluded hydrogen per cubic centimetre of manganese. These calculated values are not so large as the experimental value recorded above, but it should be borne in mind that the *isolated* Mn—H systems have been assumed to be identical with *contiguous* Fe atoms, and the difference is possibly due to this.]

‡ Neumann and Strientz, 'Monatshefte f. Chemie,' vol. 13, p. 40 (1892); vol. 12, p. 642 (1891).

§ Ewing, 'Magnetic Induction in Iron and Other Metals,' pp. 83–88.

On the cubical atom theory of Lewis and Langmuir, some insight as to the relation between magnetic property and structure of the elements may be obtained which are not inconsistent with the views expressed above. Langmuir points out that "The properties of the atoms are determined by the number and arrangement of electrons in the outside layer and the ease with which they are able to revert to more stable forms by giving up or taking up electrons, or by sharing their outside electrons with atoms with which they combine. The tendencies to revert to the forms represented by the atoms of the inert gases are the strongest, but there are a few other forms of high symmetry, such as those corresponding to certain possible forms of nickel, palladium, erbium, and platinum atoms, towards which atoms have a weaker tendency to revert (by giving up electrons only)."

According to Lewis, the most stable electron arrangement is that of the pair, and, as regards the formation of compounds, Langmuir remarks: "The pair of electrons in the helium atom represents the most stable possible arrangement. A stable pair of this kind forms only under the direct influence of positive charges. The positive charges producing the stable pair may be :—

"(*a*) The nucleus of any element.

"(*b*) Two hydrogen nuclei.

"(*c*) A hydrogen nucleus together with the kernel of an atom.

"(*d*) Two atomic kernels.

"These are listed in the order of their stability."

(*a*) represents the helium atom; (*b*) the hydrogen molecule; (*c*) the systems Pd—H, Mn—H, Fe—H, Pt—H, etc.; (*d*) the more complex ferro-magnetic systems Mn—B, Mn—P, Mn—As, etc., and the Heusler alloys. In cases (*c*) and (*d*) where one or both of the atoms have complex structure, the electrons arrange themselves in octets, which may be very much distorted. Octets may hold electrons in common with other octets, but such electrons must form one, two, or three pairs. When electrons are held in common no ionisation of the atoms takes place, and in such cases the especially close relationship between the electrons forming the pair have suggested an electro-magnetic origin of the coupling force between the atoms.* Where the chemical affinity is small, or is a differential one, as is probably the case in the unstable compounds formed during occlusion, the tendency will be for the atoms to satisfy their needs by sharing electrons.

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* A. E. Oxley, 'Roy. Soc. Proc.,' A, vol. 98, p. 264 (1921).